

Observation of Reflectance Fluctuations in Metals

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Through the study of the power spectra of a monochromatic light beam reflected by metallic mirrors, fluctuations in their reflectance is observed. The power spectra were obtained down to a factor 10^{-6} below the Standard Quantum Limit, with a dynamic range of 10^5 in the frequency and power, using methods we developed. The properties of the spectra are investigated and their dependence on the material is analyzed. The physics underlying the phenomenon is also discussed. These fluctuations provide a new window for investigating the inner workings of materials.

Reflections from surfaces, such as mirrors, are ubiquitous, and are an integral part of everyday life. In physics, studying optical properties of materials is perhaps the most powerful tool for investigating their electronic and the vibrational properties.[1–4] As such, in metals, the subject has been studied for some time, and continue to be studied actively to this day[5–7]. Properties of reflection are known to depend on the wavelength of light, temperature and the material[1–4, 8], and can further depend on geometric aspects of the material, such as its size and thickness. However, fluctuations inherent in mirrors, on which we report here, seem not to have been studied so far. The problem we address may be phrased from a different intriguing perspective — can an ideal mirror yield a “perfect” reflection? Reducing this question to its simplest concrete form, if we shine a monochromatic light on an ideal metallic mirror, can we tell whether the light has been reflected or not, just from the properties of the light itself? If so, can we tell by what material? The answers we find are positive for both questions. The underlying reason is that the reflection is caused by microscopic degrees of freedom, such as electrons and ion cores[3, 4]. All these degrees of freedom fluctuate both thermally and quantum mechanically, so that they affect the light, at some level. This effect should in principle be detectable, though the question remains whether it is possible within practical limits. While the fluctuations are indeed small, we have measured the fluctuations in the reflectance in metallic mirrors and found their properties to depend on the material. This opens another window into the inner workings of metals.

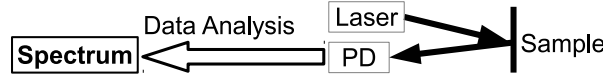


FIG. 1: Basic concept of the experiment: Laser light is shone on the sample mirror and the light reflected by it is detected by a photodetector (PD). The output current of the photodetector is analyzed and its power spectrum is computed.

When a monochromatic light beam with a constant power is shone on a flat metallic mirror, can we find the effects of the reflection in the reflected light itself? Its color is unchanged so that we essentially have only the reflected power as the property of the reflected light. However, its power can depend on time, and if microscopic degrees of freedom contributing to the reflection fluctuate, they should appear in this time dependence. To measure these fluctuations, conceptually, a simple experiment can be set up as in Fig. 1. Light is shone on a mirror and its reflection by the mirror is detected by a photodetector (PD). The power spectrum of the fluctuations in the reflected light power is $S(f) = \int_{-\infty}^{\infty} d\tau e^{-i2\pi f\tau} \langle \mathcal{P}(t)\mathcal{P}(t+\tau) \rangle$, where \mathcal{P} is the power of the reflected light, measured by the photodetector, and $\langle \dots \rangle$ indicates averaging. Fluctuations in the reflectance is $S_R(f) = S(f)/\bar{\mathcal{P}}^2$, where $\bar{\mathcal{P}}$ is the average power of the reflected light. However, measurements from such an implementation are dominated by the shot noise[9–11], the random power fluctuations in light due to its quantum nature, often referred to as the “Standard Quantum Limit”. It is impossible, even in principle, to separate the signal from this noise, with this kind of simple setup. The shot noise appears in the same manner both for the source and the reflected light, so that no effects of the reflection is observed in the light itself, with this method.

To uncover the effects of reflection, several obstacles need to be overcome. First, unwanted noise, including shot noise, needs to be reduced to levels so that the fluctuations caused by the reflection become visible. Second, it needs to be established that the observed signal is not caused by the light causing changes to the mirror itself, such as damaging its surface. Third, the cause of the observed phenomenon needs to be distinguished from other possible sources of fluctuation, such as surface waves of the material[12, 13].

To this end, the setup in Fig. 2 was used in this work. Two laser sources with wavelengths 488nm (Sapphire 488, Coherent, USA) and 532nm (Samba, Cobolt, Sweden) were combined into a single beam with a dichroic mirror (DM1), then split into two beams by a beam splitter (BS). The beams were reflected at two locations of the mirror at nearly normal incidence (separation $77 \mu\text{m}$). The light beams were focused at the mirrors down to the diffraction limit, using a microscope objective lens (Olympus MPLFLN100XBDP) with a high numerical aperture value (NA= 0.9). The

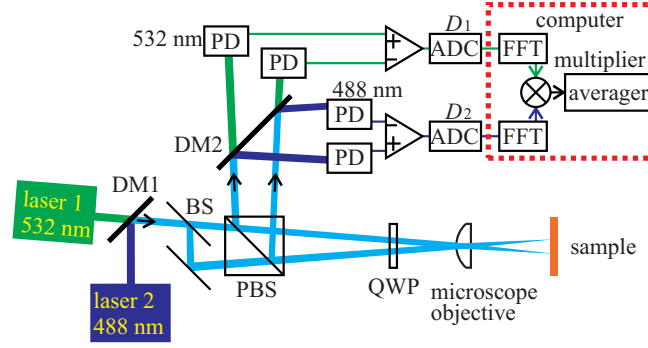


FIG. 2: Experimental configuration: Differential measurements and averaged correlations are combined to reduce the unwanted noise, such as the shot noise and the laser noise, in the measured spectra. DM1,2 transmit light 1 and reflect light 2. Paths for the laser light 1,2 are in green, blue, respectively and paths common to laser light 1,2 are in cyan. (FFT: fast Fourier transform).

reason for this is explained below. To separate the incoming and the reflected light, the light beams were circularly polarized and a quarter wave plate (QWP) was used, along with a polarizing beam splitter (PBS). The reflected light powers of the beams were measured by photodetectors (PD, Hamamatsu Photonics S5973-02).

Differential current from the two photodetector measurements using light from the same laser system (but reflected at different locations) are taken and are digitized by an analog-to-digital converter (ADC, Picoscope ps6404A), to obtain D_1, D_2 . Their Fourier transforms \tilde{D}_1, \tilde{D}_2 are computed and their correlation $\overline{\tilde{D}_1 \tilde{D}_2}$ is averaged to obtain the photocurrent power spectrum. The Fourier transforms and the averagings are performed on a computer. The principle behind the noise reduction works as follows[14]: Measurements D_1, D_2 contain both the signal S , and the noise N_1, N_2 , so that $D_j = S + N_j$ ($j = 1, 2$). For noises N_j that are independent in D_1, D_2 , $\langle \overline{\tilde{D}_1 \tilde{D}_2} \rangle \rightarrow \langle |\tilde{S}|^2 \rangle$, in the limit of infinite number of averagings, \mathcal{N} . The measurement system functions by combining the differential measurements with the averaged correlations. The former removes the light source noise, which is the same, since the source is the same. The latter reduces any noise that arises independently in the two detector measurements D_1, D_2 , such as the shot noise, statistically.

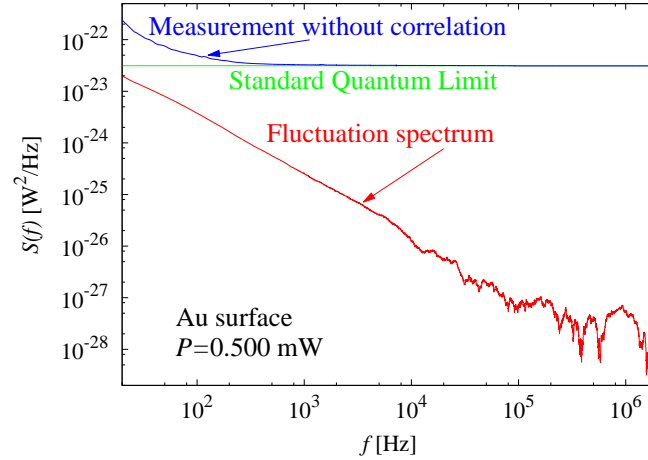


FIG. 3: A typical measured fluctuation spectrum (gold surface, $P = 0.5$ mW). Without using averaged correlations, (*c.f.* setup in Fig. 1), the spectrum (blue) is dominated by the shot noise, or the standard quantum limit (green). Making use of averaged correlations, the fluctuation spectrum (red) was obtained down to levels 10^{-6} times the shot noise level.

Results from such a measurement is shown in Fig. 3, where it is seen that the signal was measured down to 10^{-6} times the Standard Quantum Limit, with around 10^5 factor in the dynamic range, both in the frequency and the spectrum magnitude. The spectra were normalized using the shot noise level[9–11], $2eI$, in the photocurrent power spectrum, where I is the photocurrent and e is the electron charge magnitude. The spectra $S(f)$ were normalized for the output signal for a single photodetector and the reflectance fluctuation spectra $S_R(f)$ are independent of the normalization. The light beam powers applied were $2 \mu\text{W}$ to 5 mW at the mirror. Metal coated planar mirrors of

unprotected gold (PF10-03-M03, Thorlabs, USA), protected aluminum (TFAN-15S03-10, Sigma Koki, Japan), and protected silver (PF10-03-P01, Thorlabs, USA) were used in the experiment.

The light beams in the experiment travel through and are reflected by various materials, including beam splitters, a quarter wave plate, dichroic mirrors, a lens and air, apart from the sample mirror. Therefore, it is imperative to establish that the measured fluctuations arise from the reflections by the sample mirror at the two beam spots. The underlying physics for this is that the beams are focused down to the diffraction limit only at the mirrors, so that the fluctuations from other components are averaged out over the beam. This is why an objective lens with a large numerical aperture ($NA=0.9$) was used to focus the beam to its diffraction limit at the mirror, and another reason why a setup as simple as Fig. 1 is insufficient. The cause of the fluctuations was also experimentally confirmed as follows: During measurements, the light beams from the two light sources were focused on the same beam spot. When the beams were focused on different points, while keeping the rest of the light paths still overlapped, the fluctuation spectrum disappeared, showing that the fluctuations originate from the reflections at the sample mirror.

Several measurements were made at different locations of the mirror to confirm the reproducibility of the data in each case. The measurement times for the spectra were 3×10^4 s to 3×10^5 s, with more averagings, and hence longer times required for lower light powers.

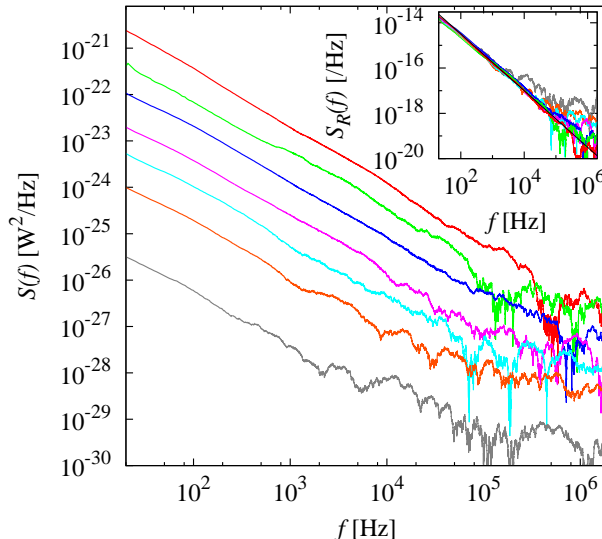


FIG. 4: Measured reflection fluctuation spectra $S(f)$ for the gold surface for light powers, $\mathcal{P} = 5.00$ mW (red), 2.54 mW (green), 1.08 mW (blue), 500 μ W (magenta), 274 μ W (cyan), 124 μ W (orange), and 19.4 μ W (gray). Spectral magnitude increases with \mathcal{P} . (Inset) The same spectra divided by \mathcal{P}^2 are seen to be identical within experimental uncertainties, validating its interpretation as the reflectance fluctuation spectrum. A fit $10^{-12} \times f^{-1.25}$ (black) is shown and is seen to agree with the reflectance fluctuation spectra nicely.

The observed signal does not exist in the incoming source light and is the sign of reflection by the mirror. However, more work is needed to ascertain whether this is a property of the material or the light affecting the mirror. To this end, power spectra for the reflected light were measured for different incoming light beam powers. The results are shown in Fig. 4 for a gold mirror. One clearly sees that the spectra are similar in shape, which indicates that the light is acting as a probe and is not affecting the mirror in an essential manner. Had the light affected the mirror, it is difficult to imagine that the spectral shape is unaffected, since the effects should grow with the power of the light beam. Also, if the intrinsic behavior of the mirror is being observed, the process should be linear, so that the power spectrum should be proportional to the square of the average light power, \mathcal{P}^2 . This can be seen in Fig. 4(inset); by rescaling the spectra by \mathcal{P}^{-2} , the spectra essentially become identical, showing the similarity of their shape and its dependence on the light power as \mathcal{P}^2 . The frequency dependence of the spectrum is well described over the whole measured range by $f^{-1.25}$ (f frequency), which can also be observed from the spectra in Fig. 4. To distinguish these fluctuations from traveling waves on the mirror surface, previously measured in light scattering at non-specular directions[12, 13], the spectra were measured using differences in the light power fluctuations at two close locations, separated by 77 μ m. Surface waves with longer wavelengths will be correlated and eliminated in this differential measurement.

A natural question is what happens for other metals. In Fig. 5, the power spectra of the reflected light are shown for an aluminum mirror for various light beam powers and in Fig. 5(inset), the spectra rescaled by \mathcal{P}^{-2} are shown. It is again seen that the spectral shapes are essentially independent of the light power and their magnitudes behave as

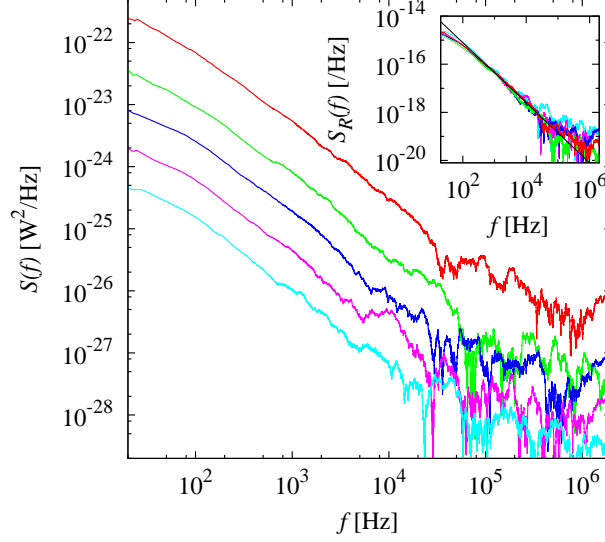


FIG. 5: Measured reflection fluctuation spectra $S(f)$ for the aluminum surface for light powers, $\mathcal{P} = 2.54$ mW (red), 1.08 mW (green), $500 \mu\text{W}$ (blue), $274 \mu\text{W}$ (magenta) and $124 \mu\text{W}$ (cyan). Spectral magnitude increases with \mathcal{P} . (Inset) The same spectra divided by \mathcal{P}^2 are seen to agree, similarly to those for the gold mirror. A fit $2.5 \times 10^{-13} f^{-1.25}$ (black) is shown and is seen to agree with the reflectance fluctuation spectra for higher frequencies, but a slight crossover behavior is seen at $f \sim 200$ Hz.

\mathcal{P}^2 . The frequency dependence is again seen to be well described by $f^{-1.25}$, but there is a slight crossover behavior at around 200 Hz. The frequency dependence of the spectra could have been different for different metals, but interestingly enough, were similar for gold and aluminum, except for the crossover behavior that exists for aluminum at low frequencies. We also measured the power spectra for a silver mirror and found that their shapes are independent of the power and consistent with the $f^{-1.25}$ behavior.

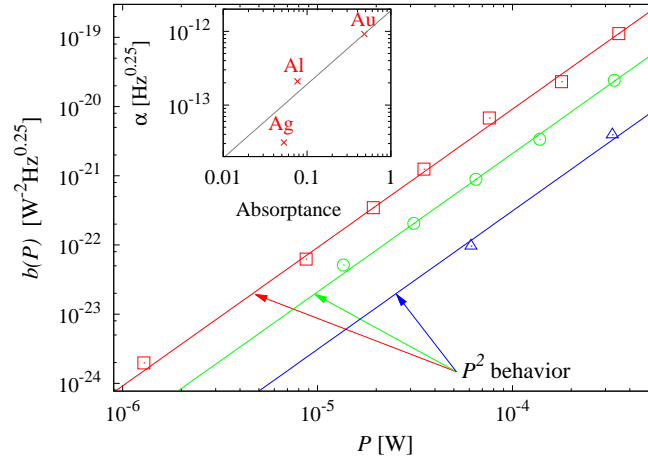


FIG. 6: The dependence of the magnitude of the spectra, $b(\mathcal{P})$, on the average light beam power, \mathcal{P} , for gold (\square , red), aluminum (\circ , green) and silver (\triangle , blue). Fits to the data for \mathcal{P}^2 behavior are also shown, and are seen to fit the data well. (Inset): Dependence of the reflection fluctuation spectra of the mirror on the material, for gold, aluminum, and silver (see text). Behavior proportional to the absorbance is also shown (gray), for reference.

To quantitatively analyze the spectra, we express the spectra as $S(f) = b(\mathcal{P})f^{-1.25}$, for $f \gtrsim 200$ Hz. The behavior of $b(\mathcal{P})$ with respect to the light power is shown in Fig. 6. It is found that $b(\mathcal{P})$ does depend on the light power as $b(\mathcal{P}) = \alpha \mathcal{P}^2$, as mentioned above, and α depends on the metal. To understand the underlying dynamics, the dependence of the α on the absorbance, $A = 1 - R$ (R : reflectance), is shown in Fig. 6(inset). α is seen to be roughly proportional to the absorption rate.

For optical wavelengths, the absorbance decreases with increasing free electron density[1–4], and since the statistical

noise from independent objects decrease with their number, perhaps this suggests the source of the spectra. Free electrons play a most important role in reflection, and it might seem that they are responsible for the observed spectra. This is, however, unlikely for the following reasons: First, the time scales corresponding to the observed spectra are in the range 0.1 s to 0.1 μ s. Free electrons in these metals have mean free times in the order of 10^{-14} s and transit times in the light beam of 10^{-12} s order. The time scales for free electrons are therefore too short to generate non-trivial correlations to appear in the spectra at the observed frequency range. Second, the energy of the photons in the light beam are 2.4 eV which is of the same order as the work function for these metals of $4 \sim 5$ eV. Therefore, observation of the fluctuations in the free electrons is expected to be non-passive and lead to nonlinear behavior. This is inconsistent with the linear behavior seen in the spectra, as in Fig. 6. Since the time scales are so short for free electron processes, these effects should not show up in the observed frequency range, though collective motions with the appropriate time scales might be able to explain the observed phenomenon.

One possible source of the observed fluctuations is the thermal motion of the ion cores in the metal, in the light beam, down to the skin depth. The number density of ion cores is identical to that of the free electrons, up to the valency factor. The time scales of their thermal motion are much longer than the time scales of free electron motion, and the ion cores do not move out of the beam so that correlations arise within the observed time scales. While the reflection is mainly caused by free electrons, their fluctuations are not observable in the correlations in the range of times that were observed here so that the ion core contributions can dominate the observed fluctuation spectra. The spectra measured in this work have behaviors close to $1/f$ over a wide frequency range, and belong to a class often referred to as the “ $1/f$ noise”, which is widely seen in nature[15, 16]. In metals, thermal motion of atoms, including the effects of internal friction is known to lead to an $1/f$ spectrum, with additional frequency dependencies coming from the frequency dependency of the loss angle.[17–20] These motions modulate the frequency of the light through Doppler shifts, which appear in the spectra, similarly to selective reflection[21, 22]. The loss angle values are not known with precision[23] and the mechanism is technically involved.

More work needs to be done to clarify the dynamics behind the reflectance fluctuations we have observed, both qualitatively and quantitatively. Measurements performed at different wavelengths can lead to more information, especially since the reflection mechanism depends on the wavelength of light[2–4]. Fluctuations in the reflectance of metals yield another window into the mechanism underlying reflection, and understanding them would lead to deeper insight into the inner properties of metals. Similar measurements can be performed for different metals, other types of mirrors and various surfaces. How the spectrum depends on the material would be an intriguing question, and even more so, why.

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- [1] P. B. Johnson and R. W. Christy, Optical Constants of the Noble Metals, *Phys. Rev. B* 6, 4370–4379 (1972).
 - [2] E.D. Palik, *Handbook of Optical Constants of Solids*, Academic Press (New York, San Diego, 1997)
 - [3] J.M. Ziman, *Theory of Solids*, Cambridge University Press (Cambridge, UK, 1972).
 - [4] N.W. Ashcroft and N.D. Mermin, *Solid State Physics*, Saunders College (Philadelphia, USA, 1976).
 - [5] R.L. Olmon, B. Slovick, T.W. Johnson, D. Shelton, S-H. Oh, G.D. Boreman, and M.B. Raschke, *Phys. Rev. B* 86, 235147 (2012).
 - [6] H.U. Yang, J. D’Archangel, M.L. Sundheimer, E. Tucker, G.D. Boreman, and M.B. Raschke, *Phys. Rev. B* 91, 235137 (2015).
 - [7] D. B. Tanner, *Phys. Rev. B* 91, 035123 (2015).
 - [8] K. Ujihara, *J. Appl. Phys.* 43, 2376 (1972).
 - [9] W. Schottky, *Ann. Phys.* 57, 541 (1918).
 - [10] S. O. Rice, *Bell Syst. Tech. J.*, 23, 282 (1944); 24, 46 (1945).
 - [11] R. Loudon, *The Quantum Theory of Light*, Oxford University Press (Oxford, UK, 2000).
 - [12] J.R. Sandercock, *Solid State Commun.* 26, 547 (1978).
 - [13] J.G.Dil, *Rep. Prog. Phys.* 45, 285 (1982).
 - [14] T. Mitsui, K. Aoki, *Phys Rev E* 80, 020602(R) (2009).
 - [15] W. H. Press, *Comments Astrophys. Space Phys.* 7, 103 (1978),
 - [16] P. Dutta and P. M. Horn, *Rev. Mod. Phys.* 53, 497 (1981).
 - [17] C.M. Zener, *Elasticity and Anelasticity of Metals*, The University of Chicago Press (Chicago, 1948).
 - [18] P.R. Saulson, *Phys. Rev. D* 42, 2437 (1990).
 - [19] A. Gillespie, F. Raab, *Phys. Rev. D* 52, 577 (1995).
 - [20] F. Bondu, J.Y. Vinet, *Phys. Lett. A* 198, 74 (1995).
 - [21] M. F. H. Schuurmans, *J. Phys.* 37, 469 (1976).
 - [22] T.Mitsui, K.Sakurai, *Jpn. J. Appl. Phys.* 36, 896 (1997).

- [23] M.S. Blanter, I.S. Golovin, H. Neuhäuser, H-R. Sinning, Internal Friction in Metallic Materials: A Handbook, Springer-Verlag (Berlin, 2007).